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Raman probes based on optically-poled double-clad fiber and coupler

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Abstract: Two fiber Raman probes are presented, one based on an optically-poled double-clad fiber and the second based on an optically-poled double-clad fiber coupler respectively. Optical poling of the core of the fiber allows for the generation of enough 532nm light to perform Raman spectroscopy of a sample of dimethyl sulfoxide (DMSO), when illuminating the waveguide with 1064nm laser light. The Raman signal is collected in the inner cladding, from which it is retrieved with either a bulk dichroic mirror or a double-clad fiber coupler. The coupler allows for a substantial reduction of the fiber spectral background signal conveyed to the spectrometer.

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1. Introduction

Raman spectroscopy is a powerful technique for the rapid and non-destructive identification of the molecular composition and structure of substances. In particular, a fiber-based Raman probe is a valuable diagnostic and monitoring tool, since its flexibility and reduced size allows for in vivo analysis, even in remote or hazardous environments. Ever since the first demonstration in 1983 [1], substantial improvements in the design of fiber-based Raman probes have been reported, in the attempt to further miniaturize the devices, enhance their collection efficiency and to integrate as many of the required functionalities as possible into fiber components [2–5]. Three desired functions can be identified: (a) generation of the excitation light; (b) its delivery to the sample and the efficient collection of Raman scattering; and (c) guidance of the latter to a suitable detector. Spectral filtering and analysis are functionalities demonstrated in-fiber elsewhere, making use of fiber Bragg gratings [6], photonic crystal fibers [7] and multimode fibers [8]. (a) A fiber laser represents a good choice for the generation of the excitation light.

However, excitation wavelengths in the visible range are usually preferred for Raman spectroscopy, due to the inverse proportionality between the intensity of the Raman signal and the fourth power of the wavelength used to excite it, while monolithic fiber lasers are generally available in the near infrared (IR). Ferroelectric crystals used for frequency doubling remove some of the advantages of the monolithic design, such as low coupling loss, alignment-free operation and increased robustness. Alternatively, second-harmonic generation (SHG) can be directly achieved in fiber by poling. In this way, the all-fiber design is preserved and both the generation of fundamental and second harmonic (SH), as well as its delivery to the sample can be done in-fiber. Frequency doubling by thermal poling followed by periodic erasure allows for generation of red [9], green [10] and blue [11] wavelengths and recently as much as 236mW of green light were produced in a fiber with this technique [12]. Ultimately, thermal poling should be used for in-fiber SHG. In this work, however, optical poling [13, 14] is used. Although the peak conversion efficiency of optical poling does not exceed a couple of percent, the technique is considerably simpler to implement since it relies on the creation of a self-organized grating [15] through the interaction of high-power radiation at 1064nm with the SH light at 532nm, requiring neither internal electrodes nor periodic UV exposure. The process is accelerated by briefly seeding the fiber with SH light, together with the fundamental IR [16].

(b) Being a low-efficiency process, Raman spectroscopy requires intense excitation, facilitated by single-mode core illumination of the sample (i.e. small beam-size diameter of the pump source). On the other hand, signal collection through a large-diameter multi-mode core is significantly more efficient. From a simplicity and sensitivity point of view, Raman spectroscopy with a single-fiber geometry is preferable over a double-fiber arrangement owing to the perfect overlap between the excitation and collection light cones at all distances from the fiber tip [17]. However, single-fiber probe measurements are normally affected by a strong interference from the fiber spectral background (FSB), i.e. the fluorescence and Raman scattering induced in the fiber by the excitation light, more intense in the fiber core due to the presence of the dopant [18]. Being structured and usually orders of magnitude more intense than the Raman signal under investigation, this FSB hinders the correct detection of the Raman lines, especially those located at low wavenumbers. This can be overcome by delivering the excitation light to the sample through a hollow-core photonic crystal fiber (PCF), which generates little or no FSB [19, 20]. Hollow-core PCFs have also been used for simultaneous pump light delivery and Raman signal collection [21, 22]. An alternative to PCFs is given by coaxial double-clad fibers, which combine the advantages of single- and multi-fiber geometries by providing an intrinsic separation between the excitation and collection paths (the core and inner cladding, respectively). This in turn substantially reduces the collected FSB. Optical probes based on double-clad fibers for endoscopy [23, 24] and two-photon fluorescence detection [25] have been previously reported. Here, a coaxial double-clad fiber is used for Raman scattering studies.

(c) The single-fiber design requires the excitation light and Raman signal to be demultiplexed, so that the collected Raman light can be retrieved from the inner cladding. This task is normally performed by a bulk dichroic filter or beam splitter, though the losses introduced by such a component can overcome the gain of using a single-fiber design [2]. An alternative to using a bulk demultiplexing component is to employ a double-clad fiber coupler, able to couple the light between the inner cladding layers of two pieces of the same double-clad fiber. Double-clad fiber couplers were recently fabricated by fused biconical taper method [26–28], side-polishing [29], or by simple contact between the inner claddings of the fibers, after the removal of the low-index polymer layer that serves as an outer cladding [30].

The work here reported addresses the frequency-doubling of the excitation light, its delivery to the sample and the collection and demultiplexing of the scattered light. By optically poling the core of a double-clad fiber enough monochromatic green light is generated to perform Raman

spectroscopy on a sample of dimethyl sulfoxide (DMSO). A double-clad structure allows for the simultaneous delivery of the generated excitation light to the sample through the core and the efficient collection of the weak scattered light in the inner cladding. Moreover, a fiber coupler able to couple the collected Raman signal between the inner-cladding layers of two pieces of the same double-clad fiber is also demonstrated in a proof-of-principle experiment. The double-clad fiber coupler, fabricated by simply etching the fibers and ensuring the direct contact of the multi-mode inner cladding regions, is optically poled and thus able to generate enough visible light to excite Raman scattering.

2. Materials and methods

2.1. Double-clad fiber

The coaxial double-clad fiber geometry provides a convenient separation between the excitation and collection paths of the probe, by allowing the simultaneous monochromatic illumination of the sample from the core and the signal collection in the inner cladding, and at the same time ensuring that the entire excitation cone is contained into the collection one. The double-clad fiber used throughout this work is shown in Fig. 1. It consists of a step-index structure with a central $7.5\text{ }\mu\text{m}$ diameter germanium-doped core surrounded by a $260\text{ }\mu\text{m}$ diameter pure-silica inner cladding and by a $20\text{ }\mu\text{m}$ -wide layer of fluorine-doped outer cladding of lower refractive index. The outer fiber diameter is $300\text{ }\mu\text{m}$, protected by a conventional acrylate primary coating. The core and inner cladding numerical apertures (NAs) are 0.12 and 0.22, respectively.

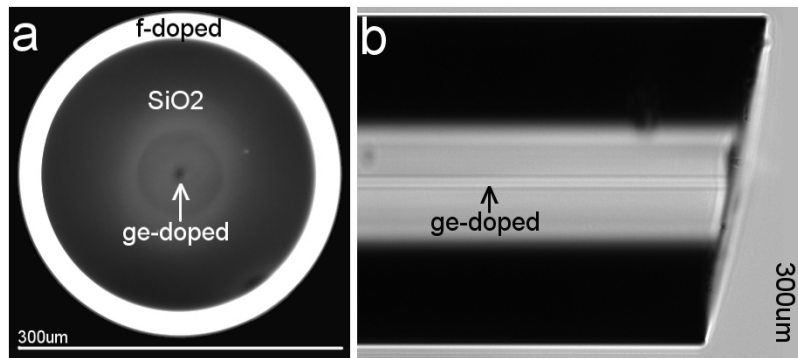


Fig. 1. (a) Cross-section of the dual-clad fiber used all throughout this work; the slightly darker circle surrounding the ge-doped core is due to the fiber fabrication process (*caning* procedure). This waveguide was specifically designed for this work. (b) Angle-cleaved tip (about 11°) on the sample side.

2.2. Optical poling

Prior to its preparation for SHG, the fiber is hydrogen-loaded for about 1 week at a pressure of 145bar at room temperature, and then stored in a fridge at -78°C to prevent any outdiffusion of the hydrogen. The H_2 -loading was reported to enhance the efficiency of the optical poling process at 1064nm [31], but it is also found to accelerate the preparation of a fiber for SHG [32]. Radiation from a Q-switched mode-locked Nd:YAG laser (Quantronix 416) is first frequency-doubled in a KTP crystal and coupled into the core of the double-clad fiber in order to generate the seed. After a few seconds the nonlinear crystal is removed and the growth of the SH continues as a self-sustained process. Saturation occurs after approx 4 hours, with $\sim 0.83\text{mW}$ average power of green light generated along the core of the fiber for 450mW IR

input. Figure 2 shows the growth of the SH as a function of time: the trend of the unseeded growth is exponential, in accordance with what is reported in the literature [13].

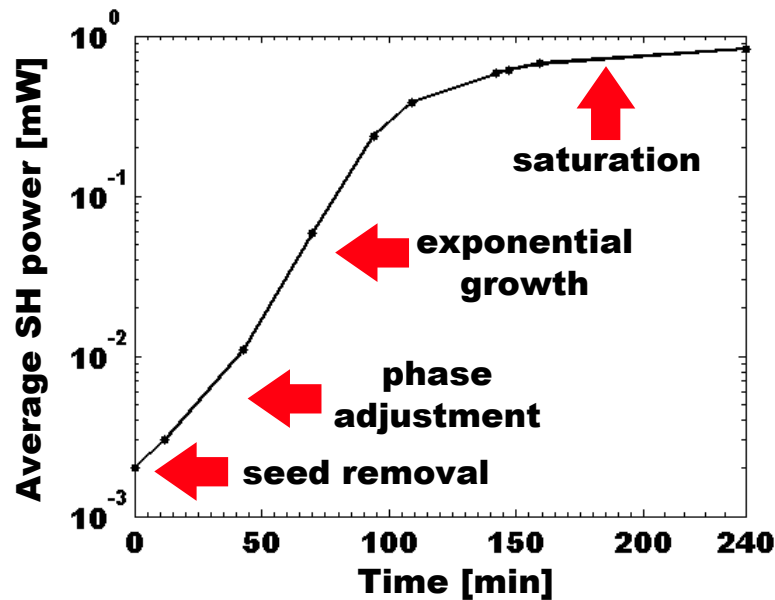


Fig. 2. Growth of the SHG over time, measured at the output of the double-clad fiber and after a 20x focusing lens and an additional IR mirror. After an initial adjustment of the relative phase of the fundamental and SH waves which follows the removal of the seed, the unseeded growth is exponential in time.

2.3. Double-clad fiber coupler

Local direct contact between the inner claddings of two 120cm-long pieces of the H_2 -loaded double-clad fiber makes optical coupling possible. To this end a 5cm-wide section of each fiber is etched in 40% concentration hydrofluoric acid for 31 minutes at room temperature. This results in $\sim 30\mu\text{m}$ -fiber radius reduction, enough to ensure the complete removal of the $20\mu\text{m}$ -wide low-index outer-cladding layer. After cleaning with acetone, the waveguides are crossed the one over the other 4 times and fixed to translational stages with UV-curable glue, as illustrated in Fig. 3. Attention is paid not to twist the fibers, in order to avoid unnecessary losses. Once the fibers are secured to the stages, axial tension is applied to ensure close contact between them, and a drop of index-matching oil (Cargille, $n_D = 1.456$) is poured over the coupling region to facilitate optical coupling. In this study, the coupler is not packaged. The required number of crossovers is determined by trial-and-error, increasing until $\sim 3\text{dB}$ coupling is achieved at 632.8nm . This is done with the aid of a HeNe laser (Melles Griot), strongly attenuated and loosely coupled (at an angle) to one port of the coupler, while the output power at the through and cross ports is monitored by means of a CCD-based spectrometer (Ocean Optics QE65000), cooled down to -20°C to reduce the dark current noise. The fourth remaining fiber tip is index-matched to avoid reflections. After the fabrication of the coupler, one of its fibers is prepared for SHG as described previously.

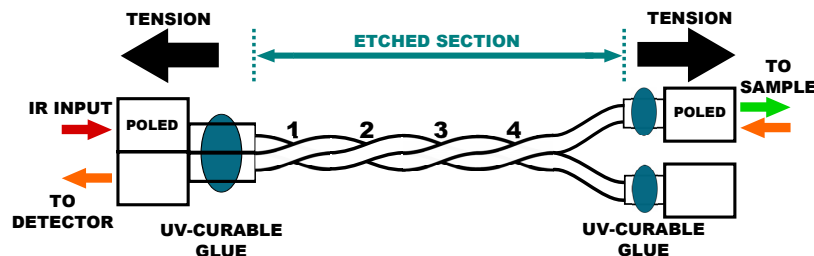


Fig. 3. Schematic illustration of the double-clad fiber coupler. The fibers are crossed the one over the other 4 times to ensure a close contact. A drop of index-matching oil (not shown) is poured over the coupler to facilitate the coupling. IR light (red arrow) is injected into the core of the poled fiber of the coupler at the end of which both the fundamental (not shown) and the SH (green) illuminate the sample. $\sim 50\%$ of the collected scattering (orange), propagating in the inner clad, couples out to the second fiber of the coupler. Rayleigh-scattered light not shown.

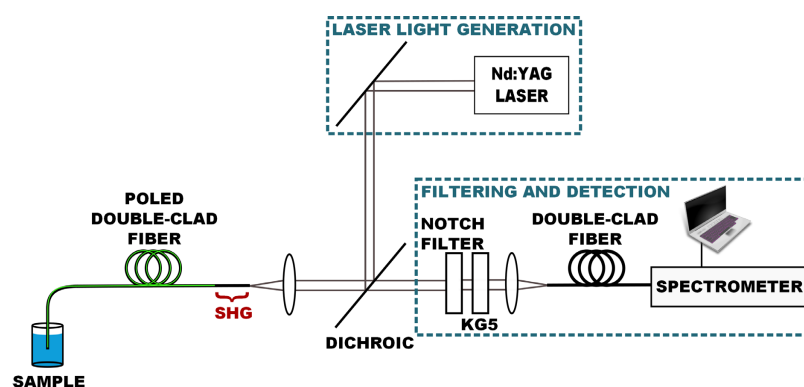


Fig. 4. Schematic illustration of the first experimental setup: SHG takes place along the core of the double-clad fiber, while the Raman-scattered light is collected in the inner cladding. A bulk dichroic mirror demultiplexes the IR radiation injected into the fiber and the collected signal.

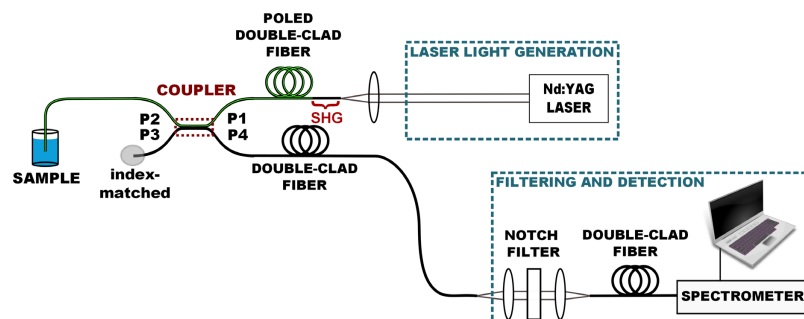


Fig. 5. Schematic illustration of the second experimental setup: SHG takes place along the core of one arm of the coupler, while the Raman-scattered light is collected in the inner cladding of the same waveguide, then couples out to the inner cladding of the second fiber.

2.4. Raman spectroscopy with double-clad fiber probe

The poled double-clad fiber is used to perform Raman spectroscopy of samples of DMSO (Merck), according to the two experimental setups schematically illustrated in Figs. 4 and 5,

and described in the following sections. At first, an optically-poled double-clad fiber is used in conjunction with a bulk dichroic component which recovers the scattering collected by the inner cladding (Fig. 4). Secondly, the setup is modified to accommodate an optically-poled double-clad fiber coupler, as shown in Fig. 5. The component couples the collected Raman scattering between the inner-cladding regions of two pieces of the same double-clad fiber, thus demultiplexing the counter-propagating excitation and collected signals without requiring a dedicated dichroic mirror. The mass of DMSO contained within the illuminated volume is approximately 7mg. In both setups, the radiation at 1064nm used to illuminate the poled waveguide originates from a Q-switched mode-locked Nd:YAG laser (Quantronix 416). Each Q-switched pulse is composed by about 20 mode-locked pulses of the duration of ~ 150 ps, generated at a 3.2kHz rate. The available peak power of the pulses is 60kW, assuming that all energy can be coupled into the single-mode core.

2.4.1. Double-clad fiber probe

This IR laser light is coupled to the core of the poled double-clad fiber by means of a dichroic mirror and a 10x focusing lens. The 532nm light generated along the fiber core is used to perform Raman spectroscopy of a sample of DMSO. In particular, the solvent is illuminated by the visible radiation exiting the core of the waveguide and the Raman-scattered light is collected by the inner cladding. The distal end of the fiber, cleaved at an angle of $\sim 11^\circ$ by means of a Vytran LDC400 Large Diameter Fiber Cleaver to minimize the Fresnel reflection at the glass/sample interface, is immersed vertically into the liquid. The same dichroic mirror used to couple the IR radiation into the core of the waveguide is used to demultiplex the counter-propagating waves, thus allowing for the recovery of the collected scattering, as shown in Fig. 4. The collected Raman signal is then filtered by a dual 532 and 1064nm notch filter (Edmund Optics), to eliminate the Rayleigh-scattered light and the IR light reflected by the other optical surfaces. Additional filtering of the 1064nm light is provided by a 2mm-thick KG5 glass filter, before the collected Raman signal is coupled into another piece of the same double-clad fiber by a second 10x focusing lens, and conveyed to the spectrometer. The aperture slit of the spectrometer is 1mm-high and 50 μ m-wide, therefore limiting the collection efficiency of the setup.

2.4.2. Double-clad fiber coupler probe

The bulk dichroic component is now replaced by a double-clad fiber coupler, as shown in Fig. 5. The SH excitation light, generated along the core of one of the fibers of the coupler, propagates unperturbed toward the sample (DMSO). The Raman signal is collected by the inner cladding of the same waveguide, through which it is transmitted until it reaches the coupling region. Along this region, part of the collected signal ($\sim 50\%$), couples out to the second double-clad fiber, which delivers it to the spectrometer through the dual-notch filter, a 10x lens and another piece of the same double-clad fiber.

3. Results and discussion

3.1. Double-clad fiber probe

The optical power generated at 532nm (0.83mW average power) is demonstrated to be intense enough to perform Raman spectroscopy on a sample of DMSO. At first, a spectrum of the solvent is acquired over a 15s interval, repeated twice to avoid the appearance of artifacts. This spectrum is shown in the inset of Fig. 6. Given the intense fiber spectral background (FSB), a second spectrum is acquired over the same time interval with the tip of the fiber held outside the liquid, at about 10cm from its surface. This second spectrum is then subtracted from

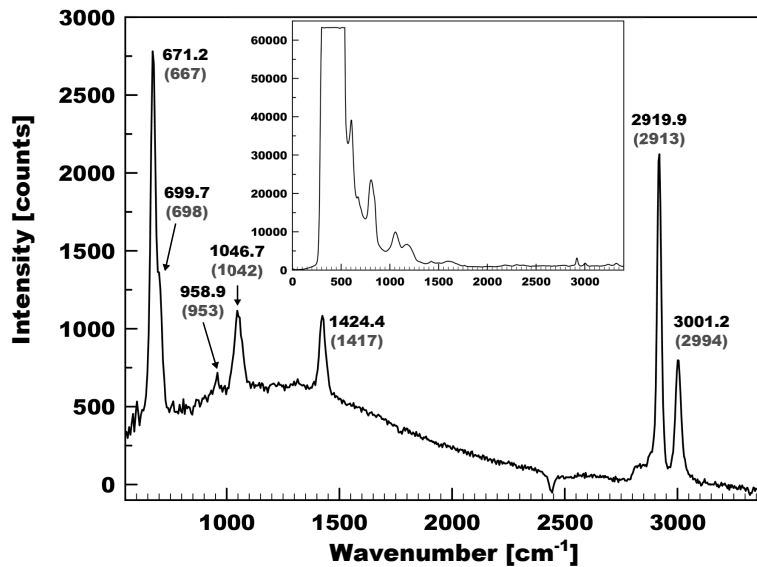


Fig. 6. Raman spectrum of DMSO acquired with the presented double-clad fiber-based probe over a 15s acquisition time repeated twice, from which the spectrum of the fiber has been subtracted. Inset: spectrum of DMSO as it appears before the subtraction. The notch appearing at about 2450cm^{-1} is due to the spectral subtraction.

the first one, and the resulting spectral difference is shown in Fig. 6. The spectral subtraction reveals the presence of the DMSO peaks located at 699.7 , 958.9 and 1046.7cm^{-1} , in addition to those appearing at 671.2 , 1424.4 , 2919.9 and 3001.2cm^{-1} , already visible in the original (unsubtracted) spectrum. These peaks are in good agreement with the work by Martens et al. [33], assumed as a reference and indicated between parentheses in Fig. 6 for comparison. The lines located at wavenumbers lower than 500cm^{-1} cannot be recovered, since in this spectral range the fiber background saturates the detector in both the spectra acquired with the tip of the fiber inside and outside the sample (see inset in Fig. 6). The FSB is mainly due to the amount of fiber background emission in the core which is backscattered and propagates back towards the dichroic mirror. This component transmits all wavelengths other than 1064nm , regardless of their origin (core or inner cladding). It is therefore reasonable to expect that the replacement of the bulk demultiplexing component with a double-core fiber coupler will considerably reduce the amount of FSB conveyed to the spectrometer, since the coupler only recovers the signal propagating in the inner-cladding region of the optically-poled waveguide.

3.2. Raman probe based on optically-poled double-clad fiber coupler

A Raman spectrum of DMSO is acquired over an integration time of 15s, repeated three times to avoid spectral artifacts. The spectrum is shown in Fig. 7. The Raman lines at 671.2 , 1424.4 , 2919.9 and 3001.2cm^{-1} are visible in the spectrum, whereas the others are hidden by the FSB signal. As expected, the background is significantly reduced with the use of the coupler. The ratio of the intensities of the DMSO peak at 2919.9cm^{-1} and the FSB peak at 802.3cm^{-1} (arbitrarily chosen as reference peaks for the two signals) in the coupler-based probe (Fig. 7) compares favorably with the case of the poled double-clad fiber without the coupler (inset of Fig. 6). The former, 0.26, is in fact twice as intense as the latter, 0.13. A second spectrum is

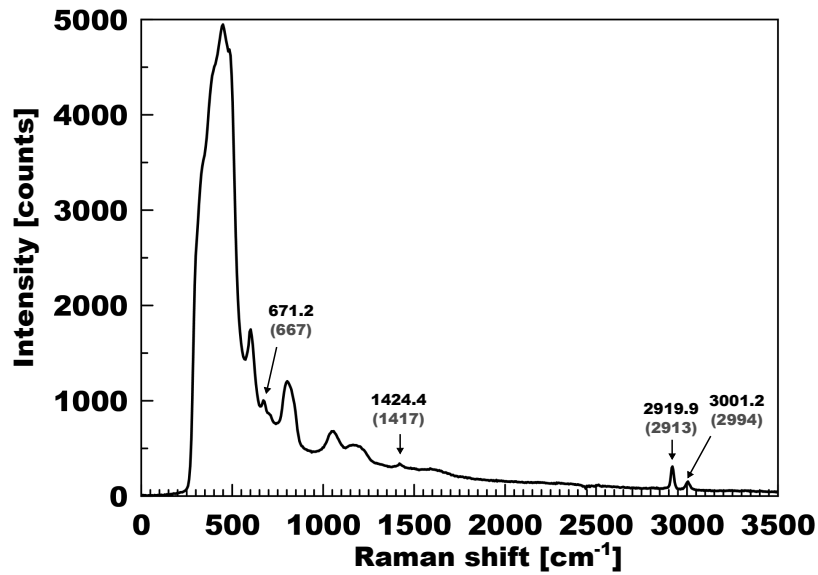


Fig. 7. Raman spectrum of DMSO acquired with the presented double-clad fiber coupler-based probe over a 15s acquisition time repeated three times.

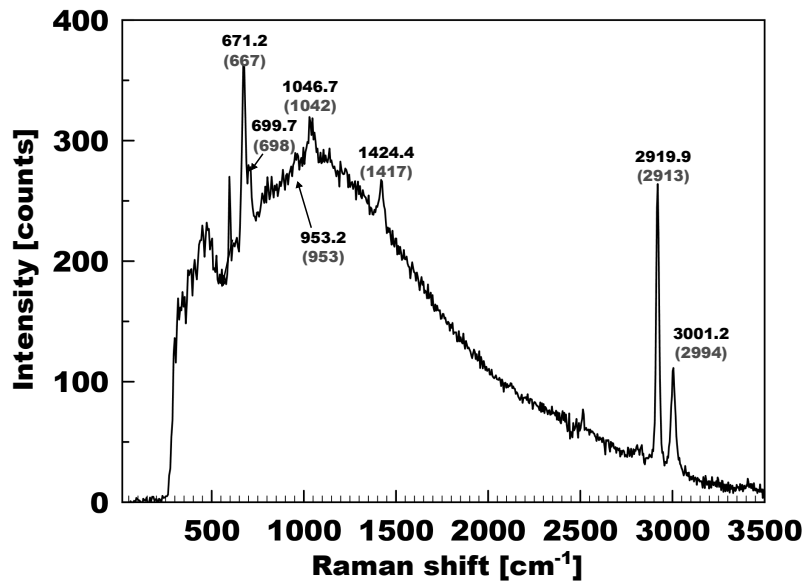


Fig. 8. Raman spectrum of DMSO acquired with the presented double-clad fiber coupler-based probe over a 15s acquisition time repeated three times, from which the spectrum of the fiber has been subtracted.

then acquired with the tip of the fiber outside the sample, to be subtracted from the first one. This spectral subtraction, shown in Fig. 8, results in the appearance of the lines at 699.7 and 1046.7 cm^{-1} . The attribution of the line located at 953.2 cm^{-1} is uncertain, given the level of

noise in the spectrum. The entire $0 - 3500\text{ cm}^{-1}$ range is shown in the spectral subtraction, since this time the FSB does not saturate the detector. The "hunchbacked" profile of the spectrum, less noticeable but yet present even in the spectrum from Fig. 6, is due to the slightly different intensities of the recorded FSB in the measurements with and without the sample, and to the fact that this background signal is mainly affecting the short-wavelength side of the spectrum. As mentioned previously, this experiment was meant as a proof-of-principle, aiming at demonstrating that an optically-poled double-clad fiber coupler can efficiently replace a bulk dichroic component. The outcome of this experiment exceeds expectations, since not only the most intense Raman lines, but also the majority of the weaker ones appears in the collected Raman spectrum (Fig. 8). It should be noted that the Raman scattered signal under study here is typically 6-orders of magnitude weaker than the levels measured in fluorescence spectroscopy [5].

4. Conclusion

Two single-fiber Raman probes based on optically-poled double-clad fibers are demonstrated in the present work. When illuminated with 1064nm light, the poled waveguide generates sufficient green light to perform Raman spectroscopy on a sample of DMSO. The double-clad structure allows for the simultaneous delivery of the excitation light in the core and collection of the Raman-scattered signal in the inner cladding. An optically-poled double-clad fiber coupler allows for the recovery of the collected Raman signal thus replacing the lossy bulk dichroic component commonly used. The coupler allows for a substantial reduction of the fiber spectral background conveyed to the spectrometer, and represents a step ahead toward the fabrication of an all-fiber Raman system, which could comprise a fiber laser at 1064nm spliced to the poled arm of the double-clad fiber coupler, and a fiber-based Rayleigh-rejection filter to remove the elastically-scattered portion of the collected light. Improvements of the results here presented include utilizing thermally poled fibers with much higher conversion efficiency for increased signal-to-noise ratio and fabricating the coupler by means of a fused biconical taper technique and packaging it for increasing the ruggedness of the Raman probe.

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